

# ITALIAN EXPERIENCE ON AUTOMOTIVE SHREDDER RESIDUE: CHARACTERIZATION AND MANAGEMENT

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**SUMMARY:** ASR can be classified as both hazardous or not hazardous waste. However both the stringent landfill legislation and the EU objectives/legislation related to ELV treatment, will limit current landfilling practice and impose an increased efficiency of the recovery and recycling of ELVs. The present paper situates ASR within the ELV context in Italy. Primary recovery techniques recycle up to 75% of the ELV components; the remaining 25% is called ASR. Characteristics of ASR and possible upgrading by secondary recovery techniques are reviewed from Italian research contributions. The application in waste-to-energy plants, in cement kilns or in metallurgical processes is analysed, with attention to the possible environmental impacts. Pyrolysis and gasification are considered emerging technologies although the sole use of ASR is debatable, its mixing with other waste streams is gradually being applied in commercial processes. The environmental impacts of the processes are acceptable, but more supporting data are needed and the advantage over (co-)incineration remains to be proven.

## 1. INTRODUCTION

Throughout the European Union an increase of 14 million tonnes of end-of- life vehicles is projected by 2015 as the number and average weight of vehicles increases (GHK/BioIS, 2006).

Directive 2000/53/EC on ELV is aimed at minimizing the impact of the end of life of vehicles on environment, by defining procedures in terms of the design of vehicles, requirements for collection and treatment facilities and attainment of targets for reuse, recycling and recovery.

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The ELVs treatment process include the de-pollution step (e.g. removal of tires, battery, lubricants and fuel) and the reclaim of spare parts and recyclable materials. Once the ELV has been processed, it may then be shredded and sorted to recover valuable metals that are recycled in iron and steelmaking processes (Nourreddine, 2007). The recovery techniques recycle up to 75% of the ELV components. The residual fraction, about 25%, is an heterogeneous material defined Automotive Shredded Residue (ASR).

Although recycling and recovery of ELV components are increasing, the ever growing number of vehicles will determine a further rise of automotive shredder residue (ASR) generated for years to come. Landfilling is currently the most widely used solution for this type of residue (Kurose et al., 2006), but due to its complex matrix the final deposit requires great technical and environmental care. Moreover, ASR landfilling is in conflict with European Directive 2000/53/EC on End-of-Life Vehicles (ELV). The Directive requires that starting from January 1st 2015: a minimum of 95% (by mass) of all end-of-life vehicles built after 1979 shall be reused and recovered, and that within the same date a minimum of 85% (by mass) of them shall be reused and recycled. In order to meet these targets on “reuse and recovery” and “reuse and recycle” ASR must be subjected to some form of recovery, since it constitutes more that 20% of the car mass.

Given the characteristics of ASR (see section 3), this is even more true when considering the Italian legislation on landfills, which forbids the disposal of waste with a Lower Heating Value (LHV) exceeding 13 MJ/kg.

A growing alternative is represented by thermal treatment with recovery of energy, which however presents the disadvantage of the toxicity of the gaseous emissions produced (Vermeulen et al. 2011). This paper focus on recent Italian research experiences on the different aspects of ASR management.

## **2. PRODUCTION OF ASR IN ITALY**

End-of-life-vehicles can result at least from two different sources: damaged vehicles and old ones (Morselli et al., 2010). Table 1 shows the number of circulating vehicles in Italy as differentiated by age and fuel feeding. A typical Italian shredder plant consists of a hammer mill, that reduces the ELV into pieces, followed by a cyclone that separates the light, non-metallic fraction (light ASR or car fluff) from the heavy one and the residual metals (heavy ASR). Afterwards, a magnetic separator recovers iron scraps from the heavy ASR and the remaining material is sent to other separation systems (such as sink-floating or Eddy current separator, which are induction sorting systems) in order to recover valuable non-ferrous metals. Table 2 shows the geographical distribution of shredding plants and processed vehicles in Italy.

Table 1. Vehicles circulating in Italy by age (ENEA 2011)

Age years	2000			2005			2009		
	Petrol	Diesel	Total	Petrol	Diesel	Total	Petrol	Diesel	Total
0-1	1.461.835	755.791	2.217.769	941.162	1.353.625	2.294.815	1.265.869	950.977	2.216.894
1-2	1.692.516	725.706	2.418.366	1.281.049	1.413.219	2.384.914	1.086.676	1.131.132	2.217.942
2-3	1.868.119	562.962	2.431.152	1.196.016	1.183.836	2.380.105	1.121.718	1.443.943	2.565.723
3-4	1.987.936	428.965	2.417.049	1.355.538	1.040.363	2.396.144	989.642	1.397.792	2.387.514
4-5	1.424.895	277.457	1.702.407	1.577.890	911.092	2.489.171	936.668	1.329.606	2.266.314
5-6	1.503.633	158.488	1.662.139	1.614.170	835.513	2.449.865	946.961	1.331.746	2.278.974
6-7	1.462.454	128.087	1.590.562	1.614.107	687.197	2.301.435	1.148.529	1.087.963	2.236.704
7-8	1.461.868	116.355	1.578.269	1.754.784	523.973	2.278.819	1.278.286	934.928	2.213.432
8-9	2.044.041	149.078	2.193.354	1.837.102	391.609	2.228.838	1.454.238	793.702	2.248.100
9-10	1.894.351	90.946	1.985.355	1.259.418	245.244	1.504.709	1.456.873	705.066	2.162.095
10-11	1.794.618	104.106	1.898.814	1.177.742	134.942	1.416.006	1.314.152	536.361	1.850.627
11-12	1.595.867	163.386	1.759.370	971.315	103.197	1.280.955	1.352.765	383.271	1.736.067
12-13	1.239.549	201.952	1.441.581	1.082.201	86.509	1.168.737	1.339.197	271.213	1.610.511
13-14	918.486	205.641	1.124.189	1.320.783	102.562	1.423.427	805.824	156.030	961.889
14-15	723.029	166.671	889.762	982.704	56.432	1.039.156	762.203	82.697	844.916
15-16	584.729	144.819	729.596	846.796	61.787	908.612	651.539	60.830	712.382
16-17	467.587	121.978	589.614	696.238	88.541	784.842	562.518	49.353	611.886
17-18	404.325	73.681	478.050	503.631	107.691	611.349	660.895	59.170	720.105
18-19	334.356	69.918	404.326	360.701	107.913	468.628	487.915	39.797	527.730
19-20	309.944	55.020	365.010	272.817	88.056	360.894	429.752	45.357	475.129
> 20	2.601.386	96.502	2.707.081	2.201.216	287.592	2.496.064	2.944.942	574.832	3.526.856
Tot.	27.775.524	4.797.509	32.583.815	24.847.380	9.810.893	34.667.485	22.997.162	13.365.766	36.371.790

Table 2. Geographical distribution of shredding plants in Italy (ENEA 2011)

Geographical macro-area	Plants	Shredder Vehicles	Recovered ferrous material	ASR
	N°	tons	tons	tons
North	22	520.243	396.641	123.602
Centre	5	195.404	152.822	42.582
South	9	159.447	103.669	55.778
Italy	36	875.094	653.132	221.962

### 3. CHARACTERIZATION

ASR is a highly heterogeneous material, that is mainly composed of plastics (23 – 41%), rubber/elastomers (9 – 21%), metals (6 – 13%), glass (10 – 20%). The broad ranges depend on the construction year, brand, engine displacement, employed technology for the treatment of disposing vehicles, the accuracy given during the dismantling operations, type of vehicles, etc.

Depending on its origin from the post-shredding scheme, ASR can be classified into (Fiore et al., 2012):

- Light fluff: fraction generated during shredding of the hulk and separated using air classification (ca. 75% of the total ASR; 10–24% of the total ELV)
- Heavy fluff: fraction remaining after metal separation from the shredded heavy fraction (ca. 25% of the total ASR; 2–8% of the total ELV).
- A soil/sand fraction is sometimes reported separately, but is usually included as part of the heavy ASR (ca. 0–2.5% of the total ELV)..

Table 3. Composition, as wt%, of ASR from Italian shredder plants.

<i>Material</i>	<i>Gabvagno (2001)</i>	<i>Mirabile (2002)</i>	<i>Marco (2002)</i>	<i>Fabrizi (2003)</i>	<i>Zolezzi (2004)</i>	<i>De Marco (2007)a</i>	<i>De Marco (2007)b</i>	<i>Mancini (2010)</i>	<i>Morselli (2010)</i>	<i>Ruffino (2010)</i>	<i>Ciacci (2010)</i>	<i>Mancini (2011)</i>	<i>Santini (2011)</i>	<i>ENEA (2011)</i>
fine material < 10 mm,	35	-	6.1			75	6.1	53.5		32.3		21.7	17	
fine material < 20 mm <sup>a</sup>								69.6	34.5			35.2		
foam	-		3.3	7	4.5			0.26		3.5		0.28		5.6
fabric, fibers	25	10	11	6	5	8	10.5	12.3	18	15.6	11	17	27	0.9
soft plastic,	20	41	29	22	20	7.5	27.6	0.26	7.5	21		0.47	23	
hard plastic								13.4	24.5	24.5	45	32.9		9.3
rubber/elastomers	20	21	9.7	23	38	4.1	35.1	1.4	13	6.6	15	15.9	9	
wiring/ceramic and electric material	-	3	0.7			0.4	0.7	0.44				3.5		
ferrous metals	-	-		13	6			1.1	2.5	6.9	9.5	2.9	8	59
non-ferrous metals				1				0.29				1.4		2
glass	-	16		13	16.5						15	0.09		2.9
wood	-	-	5.6	4	10			0.63	3	0.4		2.07	1	
paper, cardboard					1			0.41		0.4		0.4		
Others	9				2.2				30	18	13.4			20.3

Table 4. Content of metals in ASR from Italian shredder plants.

<i>Parameters</i>	<i>U.M</i>	<i>Maurino (2000)</i>	<i>Gabvagno (2001)</i>	<i>APAT (2002)</i>	<i>Mirabile (2002)</i>	<i>FAECO (2005)</i>	<i>Apat (2006)</i>	<i>Mancini (2010)</i>	<i>Viganò (2010)</i>	<i>Morselli (2010)</i>	<i>Ruffino (2010)</i>	<i>Viotti (2010)</i>	<i>Santini (2011)</i>
Fe	% wt		2.36-2.7		25.7				3-10		3.26		
Al	% wt		0.62-4.88						4-10		0.76		
As	mg/kg			4.17		6.42	0.01	15.9	3-6	16	3.44	6.4	1
Cd	mg/kg	80	30-70	22.9	62.5	16.32	31.3	22.2	0-30	6	15.2	6.7	9.91-21.1
Cr	mg/kg	900	150-200	149	800		136	209	100-200	300	226	353.3	73.1-102
Cr VI	mg/kg	<5		<1		0.49	<0.05	4.9	<1			5.06	<0.2
Cu	mg/kg	5600	7200-21800	25876	12000	2.59	12284	2,856	10000-60000	27	33500	6633	5980-21200
Hg	mg/kg		3	0.25		3.19	<0.05	0.213	0-0.5	0.8		0.31	0.17-0.42
Ni	mg/kg		100-150	69.2	700	76.2	260.38	257	50-100	210	111	191.1	34.8-38.6
Pb	mg/kg	2800	2980-3475	2007	2000	1495	6.191	2,458	0-5000	4000	410	1808	442-600
Se	mg/kg						<0.01	2.20				0.56	<0.1
Zn	mg/kg		4250-6650	3493	19000	4615	6833	19,439	0-15000		3140	14221	1810-6140

As it can be easily observed from Table 3, fines (0–20 mm fraction) can represent up to 70% of the total sample with a mean value of about 35% in wt. For the fine fraction, a thorough composition analysis is rarely performed, because of the very small size of the materials. Anyway, it is possible to identify glass pieces, plastics and metals, blended together with dust and dirt. The remaining fluff mainly consists of polymers, up to 45%, such as polyurethane (foam rubber), plastics and rubbers. Textiles accounts for about 10% on the total and together with polyurethane foam (PUF) are strictly related to car seats and carpeting (Morselli et al., 2010). Elemental Analysis of ASR from Italian shredder plants is shown in Table 4 while the main metals content is reported in Table 5.

The levels of contaminants such as Pb, Cu, Ni, and Zn substantially confirm similar characterization results from other international research works (Vermeulen et al., 2011), thus posing serious attention on the need to lower concentrations throughout the entire ELV recovery process. Considerable amounts of copper wire fragments, many connected to or intertwined with other materials, were observed in all the Italian samples and likely contribute considerably to copper levels. Some of the copper (wire fragments) should be recoverable, thus lowering the high Cu content in ASR.

The content of mercury in Italian ASR is typically around 0.5 mg/kg, roughly half the Hg present in the ELV before shredding (Hg is mainly used in switches) although values of up to 3,2 mg/kg have been reported (FAECO, 2005). Cd is typically found at 20–80 mg/kg in ASR, whilst Pb levels are much higher, typically 500–5000 ppm (0.05–0.5%), for chromium (Cr) typically 150–800 mg/kg, for arsenic (As) up to 16 mg/kg. Dealing with the total metal content, an increase of Pb and Zn can usually be observed towards the finest fraction whilst the other metals considered do not exhibit such clear grain-size dependence. Notwithstanding their total metal content, Italian ASR usually does not overpass the threshold values for landfill disposal.

Table 4. Elemental composition and characteristics of ASR from Italian shredder plants.

<i>Parameters</i>	<i>Patierno (1998)</i>	<i>Maurino (2000)</i>	<i>Galvagno (2001)</i>	<i>APAT (2002)</i>	<i>Mirabile (2002)</i>	<i>Zolezzi (2004)</i>	<i>Viganò (2010)</i>	<i>Ruffino (2010)</i>	<i>Mancini (2010)</i>	<i>Morselli (2010)</i>	<i>Santini (2011)</i>
Ash					36.2	27.3			44.7	28.2	35.8
C	17.5	40	37.2	48.11	44.5	47	40	46	36.3		
H	2.1	5	4.8	6.07	5.3	5.8	5	5.89	4.67		
N	0.5	1	-	-	4.5	1.2	2	1.68	-		
O	17.4		1.5	-	6.9	19.5	2.5		-		
S	0.25	0.95	0.4	0.3	0.2	0.3	0.3	0.4	0.28	6	0.23
Cl	0.05	3.5	2	-	0.5		1.25		0.94		0.95
F	-		-	-	-	-	0.75		0.017	<0.05	

Table 5. Other critical contaminants in ASR from Italian shredder plants.

Parameters	U.M.	ANPA (2006)	Mancini (2010)	Santini (2010)	Morselli (2010)	Viotti (2010)
IPA	mg/kg s.s.	5.8±2.7	5.6±1.6	4-6.4	4.4	3.18±1.07
PCB	mg/kg s.s.	44.45±34.06	7.9±1.7	0.008-0.11	5.3	2.97±0.39
PCDD/PCDF	ngTE/kg s.s.	56.75±44.64	3.02±0.50	3-4		1.19±0.05
Mineral oils	g/kg		22.3±3.5		26.8	

#### 4. SECONDARY MATERIAL RECOVERY

ASR is generally made of about 20-30% -wt of plastics (rigid plastic, polyurethane foam - PUF, textiles), 15-20%-wt of rubber (simple, textile/metal reinforced), 20-40%-wt of paper and wood, and of about 10% -wt of not combustible materials (i.e. inerts, such as glass, paint, soil) and metals (magnetic, non-magnetic and PVC wrapped wires) (Kim and Young, 2004; Lanoir et al., 1997; Mirabile et al., 2002; Forton et al., 2006).

The main issues in secondary raw materials recovery concern two aspects: first of all the fulfilment of the qualitative specifications that make their reuse favourable if compared to virgin raw materials, and obviously the minimization of undesirable components; afterwards, a constant demand of the secondary raw materials, involving a selling price adequate to cover the recovery process costs, should exist.

Excluding fines ( $d < 20-30$  mm, which may account for the 40-60% w/w of ASR) (Fiore et al., 2012; Morselli et al., 2010; Cossu and Lai, 2012; Mancini et al., 2010), and taking into account only the ASR coarse fraction, numerous materials may theoretically be recovered and recycled, if proper technologies are applied and financially competitive markets are available.

The technologies that may be employed to mobilize valuable components from ASR include mechanical processes (mainly shredding), and physical operations such as dimensional, magnetic, electrostatic and densimetric separation phases.

Considering glass, there does not currently appear to be any process that would be able to substantially recover this material other than in pre-shredding operations; actually glass is largely found both in the fine and in the coarse fractions.

##### 4.1. Metals recovery

ASR contains small metal pieces, both ferrous and non ferrous ones, that are left behind by the shredding process: the recovery of these components, having a high economic convenience, is usually performed in shredding facilities by means of magnetic, electrostatic and densimetric post-shredding phases (see Figure 1), allowing the recovery of a fraction equal to about the 5-6% w/w of an ELV performing the post-shredding process on real scale (Fiore et al., 2012).

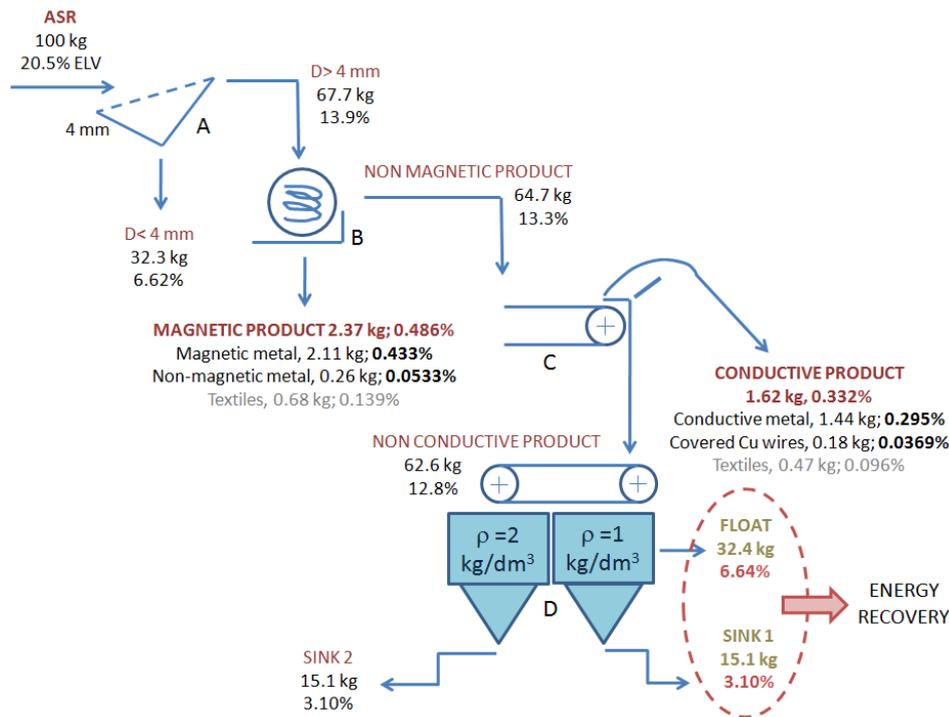


Figure 1. A possible treatment process for the valorization of metals from ASR (adapted from Ruffino et al., 2010)

## 4.2. Plastics recovery

Plastics constitute a significant fraction of ASR, but would have to be separated into individual types, or groups of types, for a reuse in specific applications. Up to 27 different types of polymeric materials are currently encountered in ASR, although about the 60% of the plastic components are made of polypropylene (PP), polyurethane (PUR), polyvinylchloride (PVC), acrylonitrile butadiene styrene (ABS) (Vermeulen et al., 2011).

According to Ferraõ and Amaral (2006a), increasing the recycling of plastics from ASR is the key issue to achieve the European reuse and recycling target of 85% by 2015. It is expected that in this way an additional 6-10% of the total ELVs mass, compared to the actual Italian average value of 78-80%, can be recycled. Industrial applications (i.e. the Galloo process in Belgium) of this solution have shown that European reuse/recycling target of 85% can indeed be fulfilled (Jody and Daniels, 2006; Vermeulen et al., 2011).

ASR amount is destined to dramatically increase worldwide in the next future: considering that in the last decades automotive manufacturers were inclined to reduce the fuel consumption of vehicles by enhancing the fraction of light components and materials, the consequence is that vehicles composition changes affect both ASR quantity and quality. The introduction of the EU-directive on ELVs moreover gave a strong incentive to manufacturers to reduce the number of different plastics used and to label plastic parts in order to facilitate the identification during dismantling pre-shredding operations, although enhancing dismantling operation also increases ELV processing costs (Ferraõ and Amaral, 2006b).

Several studies pointed out that these plastics can be physically and mechanically recovered from ASR with a sufficient purity to obtain high quality secondary raw materials (Jody and Daniels, 2006): Argonne process performs a mechanical separation of ASR to concentrate plastics and a subsequent separation of single polymeric types by means of froth flotation; RPI

International performs mechanical separation followed by skin flotation, while in Belgium Salyp process realizes the thermoplastic materials sorting. In Austria Volkswagen- SiCon process performs in Austria a multiphase process to separate plastic mixtures, ferrous and non ferrous metals. In Belgium Galloo process performs in Belgium the separation of light plastics, heavy combustible fractions (rubber and heavy plastics) and light fractions (PUF and textiles) from ASR.

Polyurethane foam (PUF), which account for about the 5% w/w of ASR and for about the 30% v/v, has a very interesting market as secondary raw material for indoor/outdoor recycled products: Argonne and Salyp processes perform the separation of PUF from ASR by screening and a successive multiple cleaning phase made of washing, rinsing, drying and baling steps (Jody and Daniels, 2006).

Robson and Goodhead (2003) granulated ASR after removing large metal parts, and tested a dual injection moulding process based on the injection in a two channels device of a core made of 45-50% of granulated ASR and 50-55% of polypropylene (PP), and a skin made of 100% PP.

The quality of the recycled plastics indeed remains a major issue, and also the elimination of certain polymeric materials, which may cause problems in some subsequent energy recovery processes, particularly pyrolysis.

Some of the most probable products for plastics recycled from ASR are goods that already achieved a satisfactory quality employing industrial waste plastics, such as outdoor equipments (i.e. containers, benches, fences). Nevertheless, considering an outdoor use of the recycled material, the elution of hazardous compounds should be prevented, and metals as undesirable components should be minimized also considering the energy recovery of combustible ASR fractions.

From this point of view, heavy metals removal may enhance the recycling/recovery potential of ASR plastics components: Kurose et al. (2006) proposed several washing processes, based on the use of NaOH or HCl or HCl/KCl solutions.

### **4.3. Fines recovery**

Shredder fines ( $d < 20-30$  mm), which may account for about the 40-60% of ASR depending on the intensity of the shredding operation, are usually made of fragments of all the ASR components. This fraction usually tends to be contaminated by oil and other fluids not carefully removed in the depollution pre-shredding phase.

Shredder fines may undergo energy recovery (Forton et al., 2006) in high energy-demanding plants such as cement kilns. Actually fines are also rich of heavy metals and contain glass, sand and other inerts, which are basic ingredients for building materials such as cement and aggregates. However the only realizable hypotheses for fines recovery as secondary raw materials should consider their inertization, keeping in mind that their hazardous contaminants may potentially pose a serious limitation.

Several studies have shown that shredder fines may be employed as building materials: Pera et al. (2004), outlining that zinc and lead, respectively accounting for the 1-3.5% w/w and for the 0.7-3.3% w/w of the fines, are ordinary retardants for Portland cement, proposed the production of aggregates either by a thermal-chemical treatment and the mixing with Portland cement, or by direct transformation in concrete by mixing with calcium sulfo-alluminate cement. The results of the performed leaching tests were positive, and particularly the direct mixing option appears very interesting.

Rossetti et al. (2006) considered the granulation of ASR fines below 4 mm with lime or cement, and tested the mixing of the obtained aggregates, which dimensions were in the 5-30 mm range depending upon water content, with sand, water and cement to produce concrete mixtures. The performed leaching tests showed the efficiency of the inertization process, leading to a total absence of metals in the leachate.

Granata et al. (2011) on the other hand explored the hydrometallurgical recovery of metals from ASR fines below 4 mm: zinc, concentrated particularly in the below 0.5 mm fraction, may be easily removed both by means of a weak acid attack with acetic acid and a strong acid attack with sulfuric acid, leading to a final recovery by electrowinning respectively of the 80% and of the 95% of the solubilized metal.

## **5. THERMAL TREATMENT**

Although EU legislation is striving towards a sharp rise in ASR recycling processes, the expected high recycling quotas (exceeding 85%), could indeed be utopian due to the very low material recovery rate of non-metals. The novel and innovative lightweight car designs, aimed at emission reduction during the usage phase, will imply a significant increase in the application of plastics and composites as well as of light metals in view of which it may prove additionally difficult in the future to achieve the required recycling quota of 85%, and even more difficult a recovery quota of 95%, by 2015 (Reuter, 2006). On the other hand, the high calorific value of ASR largely dependent on its organic fraction, (typically about 50 wt%) promote the thermal treatment methods such as co-incineration with other wastes in waste-to-energy (WtE) installations or application as (energy) feedstock in the foundry and cement industries. Whereas the small particle size of the fine ASR fraction often hinders further mechanical recovery, this fraction can largely be handled by thermal treatment thus constituting a cost effective and more sustainable alternative to landfilling of ASR.

It is however surprising as, according to the EU-Directive 2000/53/EC, the fraction of ASR that is not recycled, but from which energy is recovered, should by 2015 not exceed 10% of the original ELV's mass. Therefore, the thermal treatment has to be considered only as a complement to intensive dismantling and primary and/or secondary recovery activities. Processing of ASR through thermal treatment was however studied in various research, pilot and full scale projects and showed high potential, as ASR has a heating value ranging from 14 to 30 MJ/kg

### **5.1 Co-combustion**

The use of combustion technologies (i.e. incineration) to treat ASR may be hampered by the fact that ASR frequently contains high amounts of chlorine, heavy metals, and toxins. As a consequence, thermal treatment in Waste-to-Energy Plants (WTEP) may result in the emission of acidic gases, fine dusts containing heavy metals, and organic pollutants. In general ASR is not suited for mono-incineration, due to possible carry-over of unburned fines and melting characteristics. A mixture with lower calorific wastes enhances the incineration potential and efficiency in waste-to-energy plants, while still maintaining the advantages of considerable mass and volume reduction, along with recovery of energy (Vermeulen et al, 2011).

However, high concentrations of chlorine in ASR (up to 4 wt%) result in high concentrations of hydrogen chloride (HCl) in the combustion gases, which may reduce the service life of incineration equipment due to chemical corrosion and fouling. De-chlorination techniques for ASR are intensively researched in order to make ASR more suitable for co-incineration in installations that operate at temperatures lower than 1100 °C.

Co-incineration of ASR can be challenging also due to high concentrations of persistent organic pollutants (POPs) and heavy metals. The concentration of these components can be reduced by advanced upgrading techniques, when economically feasible. Optimum control of the process conditions in combination with adequate flue gas cleaning will limit the emission of harmful substances. In a very comprehensive review published by Vermeulen et al. 2011, it was emphasized that recent developments in sequential gasification/combustion are expected to lead

to atmospheric emissions that are well below the legal limits.

The authors also concluded that the 95% reuse and recovery target can be met by applying thermal incineration techniques or emerging technologies such as pyrolysis or gasification. These last techniques have been gradually applied in some commercial processes, especially with ASR mixed with other waste streams, but their application is still poorly documented and further research is required. The practical advantages of these techniques compared to (co-) incineration remain to be demonstrated.

## 5.2 Gassification

Gasification is commonly operated at high temperatures (>700–800 °C). Air is used as a gasification agent, and the air factor is generally 30% or 40% of the amount of air needed for the combustion of the organic fraction of the feedstock. Gasification produces mostly a gas phase and a solid residue (char and ashes). The use of air introduces N<sub>2</sub> in the gases, thereby considerably reducing the calorific value of the syngas because of the dilution. Gasification has been widely studied and applied for biomass, coal and plastic solid waste. The application for the treatment of ASR is less documented. Sequential gasification and incineration tests were reported by Mancini et al. (2010) and make use of a rotary kiln, operated between 850 and 1120 °C with an air factor <1. Combustion of the gases is completed in a secondary afterburner chamber. The system is completed with a boiler (steam at 43 bar, 430 °C) and turbine. The capacity was on average 2400 kg/h during the tests. The full-scale plant, designed for the thermo-valorization of tyres, was purpose-modified to allow for ASR combustion. Several plant components were specifically modified; for instance, the inlet of the gasification chamber was properly sealed to avoid excess air in the first reaction stage (Figure 2).

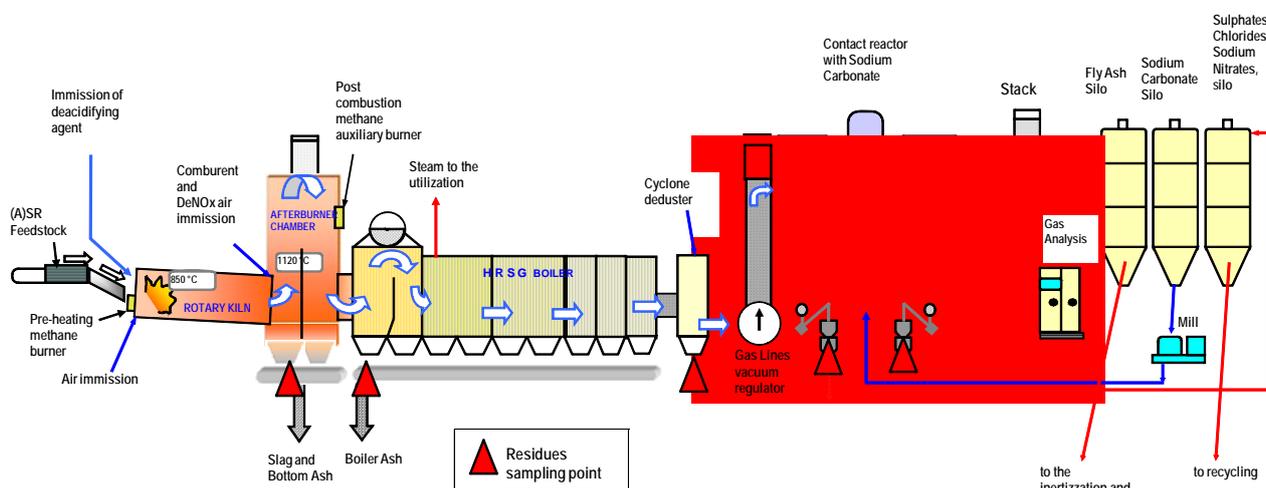


Figure 2. Layout of the gasification-combustion plant (adapted from Mancini et al., 2010)

Three different ASR compositions (ASR with different percentage of shredding, whites and 100% ASR) were taken into consideration. Both the raw samples and solid products were thoroughly characterized. Combustion emissions, continuously analyzed during the test period, were invariably considerably below the legal limits. Slag represented approximately 18% of the initial ASR by weight. Analysis of solid residues indicated that slag can be landfilled as an ultimate non-hazardous waste material while both fly and bottom ash have to be classified as

hazardous wastes (2% of ASR weight). ASR pre-treatment could be useful before gasification to increase the heat of combustion and to recover valuable metals. Preliminary results, obtained from a unsophisticated thermodynamic analysis of the process, indicated a value of 0.61 for energy efficiency parameter calculated in accordance with the Directive 2008/98/EC. The outcome of the study provides an important contribution towards assessing the feasibility and reliability of the process, thus constituting a basic prerequisite for process performance evaluation. A modelling and LCA analysis of a sequential gasification and combustion process to treat ASR, using a fluidized bed gasifier (590 °C) followed by a cyclonic afterburner (1400 °C) is described by Viganò et al. (2011). The applied technology for treatment of ASR allows the recovery of large fractions of metals as non-oxidized, easily marketable secondary raw materials, the vitrification of most of the ash content and the production of power via a steam cycle. Results show that despite the unfavourable characteristics of ASR, the proposed technology can reach appealing energy performances. Three of four environmental impact indicators and the cumulative energy demand index were favourable, the main positive contributors being electricity production and metal recovery (mainly aluminium and copper). The only unfavourable indicator was the global warming index because, since most of the carbon in ASR comes from fossil sources, the carbon dioxide emissions at the stack of the thermal treatment plant are mainly non-renewable and, at the same time, the avoided biogas production from the alternative disposal route of landfilling is minor. Operation of the afterburner at 1400 °C moreover produces a vitrified (and hence inert) slag. The authors concluded that the sequential gasification and combustion system reaches appealing energy and environmental performances, despite the unfavourable characteristics of ASR.

### 5.3 Pyrolysis

As is well known, pyrolysis is the thermal degradation of macromolecular materials in the absence of oxygen. The main products of this process are a solid phase (charcoal), a liquid phase (tar) and a gaseous phase. The conventional pyrolysis is carried out at moderate temperatures (300–600 °C) and low heating rates (with vapour residence times of 10 s to 10 min) and gives approximately equal mass fractions of gas, liquid and solid products. Pyrolysis seems to offer an environmentally attractive method for the treatment of ASR. As mentioned above, the process uses medium to high temperatures and an oxygen-free environment to decompose ASR chemically, thus producing minimum emissions of nitrogen oxide and sulphur oxide compared to the commonly practised conventional technology, incineration. Pyrolysis also allows valuable materials to be recovered. This is particularly important when designing plants to meet material recycling targets established by the ELV Directive. The process, generally, begins with pre-heating of shredded materials being fed to a reactor. In the reactor, depending upon the chosen process parameters, the waste material is heated to the required temperature. The products of pyrolysis represent a significant percent of the initial volume of organic matter that can be converted into energy, to either sustain the process or produce excess power. Processes for the pyrolysis of ASR could specifically be designed to maximize gaseous products or focus on material recovery as a key design requirement. Potential users of pyrolysis char include iron, steel and cement industries. However, these industries apply restrictions on the presence of specific metals, and chlorine mainly from PVC and other plastics, and a reduction of these components may be required either prior to or following shredding. If pyrolysed char is not recycled but forwarded to landfill disposal, knowledge of the level of metals remaining in the char is of fundamental importance to avoid problems of contamination. In designing an ASR pyrolysis process the latter problem may be avoided by using the large amounts of waste heat

available to vitrify the char. Several processes apply this solution. For the cement industry, the level of contaminant metals remaining in ASR char is generally excessive. Fundamental research of ASR pyrolysis mostly used thermogravimetric analysis and small-scale reactors as reported by e.g. di Marco et al. (2007), Zolezzi et al. (2004), Chiarioni et al. (2003). The use of pilot-scale experiments is very limited, with Galvagno et al. (2001) being the only detailed example retrieved from literature. Zolezzi et al. (2004) compared performance and product yields in conventional pyrolysis and fast pyrolysis of automotive shredded residues. The pyrolysis tests were carried out in a fed batch laboratory cylindrical reactor (inner diameter: 54 mm, length: 1000 mm), made of Hastelloy steel, with horizontal axis indirectly heated by a tubular electric furnace. The set-up for fast pyrolysis was similar to that for conventional pyrolysis. The kiln reactor was an AISI 310 steel reactor (length: 650 mm, inner diameter: 28 mm) indirectly heated by electric elements surrounding the reactor chamber. Tests were conducted in batch mode; a sample (weight 25 g). In both cases the carbon conversion was more than 80% for temperature ranging from 500 to 800 °C. Conventional pyrolysis maximised gas production (about 35% coupled with 20% of oil production), while the fast pyrolysis gave an oil yield higher than 55%. The carbon content of the conventional pyrolysis gas reached 36.4% by weight of the feed at 800 °C, while more than 69% of the initial carbon was found in the pyrolysis oil obtained by the fast pyrolysis at 800 °C. The higher heating value (HHV) of the conventional pyrolysis gas ranged from 8.8 to 25.07 MJ/Nm<sup>3</sup>. The fast pyrolysis oil HHVs ranged between 28.8 and 36.27 MJ/kg. The authors concluded as both conventional and fast pyrolysis processes offer an attractive way for the recovery of energy from ASR as an alternative to more traditional routes, such as incineration, because of the reduced environmental impact (e.g. atmospheric emissions of toxic contaminants) and the opportunity of material recovery (e.g. adsorbents from chars, chemicals from tars). Galvagno et al., (2001) carried out some preliminary pyrolysis experiments in a pilot scale installed at the ENEA research centre. The main objective of the research work was to evaluate both process performance and characteristics of the products obtained while operating the plant under different operational parameters, such as ASR load, temperature, residence time, pressure etc. ASR samples used for the treatment, solid and liquid by-products obtained and syngas produced were analysed both through on-line monitoring (for gas) and laboratory analysis. The pyrolysis unit consisted of a rotary kiln reactor (110 dm<sup>3</sup> in volume and 0.4 m in diameter) and was heated externally by using electric elements surrounding the reactor chamber. The pyrolyser operated under a positive pressure of nitrogen (300 mm w.c.). The maximum feeding rate was 60 l/h. The authors concluded as ASR gets converted into a solid residue (char), liquid fraction (oils) and gaseous fraction (syngas) and the process temperature significantly affects the relative distribution of the three products. At high temperature, up to 680°C, complete conversion of the material takes place. Moreover, the production of syngas is favoured over oil at this temperature. In addition, the process permits recovery of material from the solid residue (metal). On-site utilisation of syngas, as combustible material, decreases the overall process management costs. Although results obtained during the leaching tests conducted on char at different temperatures, revealed ash and heavy metals concentrations. double than that of the starting material, the effect was quite similar to that of ASR, i.e. the char revealed to be no more dangerous than ASR for the landfilling or for any other treatment. De Marco et al., (2007) presented an experimental study of pyrolysis of real ASR samples coming from an automobile shredder plant (light and heavy ASR). The pyrolysis experiments were carried out at 400, 500 and 700 °C in nitrogen atmosphere, using an unstirred stainless steel 3.5 dm<sup>3</sup> autoclave. In a typical run 100 g of ASR was placed into the sealed reactor. The raw samples, as well as the solid, liquid and gaseous pyrolysis products were thoroughly characterized. It was concluded that pyrolysis appears to be an appropriate technique for recycling the heavy ASR, since valuable solids (38–39%), liquids (20–29%) and gases (31–41%) are obtained, while this is not the case for the light ASR, which does not yield valuable products. Concerning temperature 500 °C was

considered sufficient to produce total decomposition of the ASR organic matter. Increasing temperature over 500 °C increases gases to the detriment of liquids, but hardly varies the characteristics of the products. A critical review of the developments in the full-scale pyrolysis of ASR has been presented by Harder and Forton (2006), giving a complete analysis of the evolution of the pyrolysis processes, and of the competing environmental, legislative and commercial drivers towards a further development. However, in spite of the numerous studies undertaken and developments made, in recent years, at various levels of commercialization, pyrolysis is still generally considered unproven for ASR treatment and few demonstrator plants have advanced to a successful commercial status. (Harder, 2007). Currently only one process is considered fully commercial—the Ebara plant in Japan (Selinger, 2003). Ebara co-processes SR with sewage sludge (70/30) at around 100,000 tons per year using gasification followed by vitrification of the residue to produce an ‘inert’ product. Three other pyrolysis processes are classified as semi or fully commercial, clearly specifying that they are authorized to handle ASR as a feed (Malkow, 2004). These are the PKA process, the Pyromelt Process (Lurgi Ensorgung), and the TWR process (Siemens; Schwel-Brenn; TWR/Mitsui). All these processes mix ASR feed with other wastes to regulate the variations in energy content and material and post-processing the char. Some use the available energy to vitrify the remaining char but this is however expensive due to associated high financial and energy costs.

#### **5.4 Other ASR fuel and feedstock applications**

ASR is a potential alternative fuel and mineral feedstock for cement production as about 50wt% of ASR consists of combustible matter such as plastic or rubber, and another 40 wt% is made up of silicates, calcium, aluminium and iron. Given these typical waste characteristics, a cement kiln seems in principle ideal for the use of ASR as fuel. However the use of ASR in the cement industry implies a need to guarantee feedstock quality parameters in order to satisfy kiln operators and certify that no significant increases in kiln emissions will be elicited (Vermeulen et al., 2011). The pretreatment and mechanical means, required to process ASR into material suitable for use as coal and mineral substitutes, should be properly assessed. Techniques aimed at reducing contaminant levels (particularly mercury) should be developed. Other problems related to co-incineration of ASR in cement kilns are increased ash formation, clogging of the fuel injection zone, and increased concentrations of hazardous elements in the cement kiln dust (Genon and Brizio, 2008). However, the use of ASR in kilns may lead to reduced environmental impacts resulting from less mining, transportation and preparation of coal and mineral ores as well and reduced impacts of SR landfill leachate. Remaining barriers to acceptance include demonstration that a full-scale system will consistently produce high value fuel with low concentrations of hazardous materials.

ASR can also be used as an alternate fuel to coal in the iron metallurgical industry with an excellent emission control (Mirabile et al., 2002). This can be attributed to the fact that blast furnaces are able to comply with the confined values fixed by national norms without any significant additional investment. Use of such material, especially in the metallurgical sector, offers energetic, environmental and economic advantages without installation of new plants. Nevertheless, the experience gained on this topic is still not sufficient to express its techno-economic evaluation. In fact, if the process on one hand is certainly efficient energetically, on the other hand, the same appears to provide end products of a modest quality.

Mirabile et al. (2002) verified in a pilot plant that temperature and atmosphere characterising blast furnaces allow the complete destruction of every organic compounds and avoid the formation of micro-pollutants. Furthermore, heavy metals are dissolved in the hot metal and in the slag. Technical and economical evaluations showed encouraging results in the range of 1–

3% by weight of ASR in the auxiliary combustible.

### 5.3 Combustion residues

Another important environmental concern related to combustion and co-combustion of ASR is the increasing concentration of heavy metals in the solid residues. As already mentioned in Section 3, automotive shredder residue is rich in certain heavy metals, such as copper, nickel, lead and zinc. Before incineration, the concentrations of heavy metals can be reduced by the advanced secondary recovery techniques. Especially the high amount of copper present in ASR, mainly transferred into the bottom ash, may limit the application of this ash as a raw material. The fly and boiler ashes are most often landfilled together with the FGC-residue, possibly after an appropriate treatment if required. Especially the higher concentrations of Cd, Pb and Zn might require removal or stabilization of these heavy metals before landfilling.

Recovery of bottom ash and slag after ASR thermal treatment (co-combustion, gasification/combustion) is an option usually unevaluated in most countries (e.g. Italy) due to excessive amounts of contaminants, especially metals.

Various metals (especially iron, aluminum, copper, zinc, lead) can however be removed and recovered from ASR incineration bottom ash and slag by a combination of mechanical (e.g. crushing, sorting) and physical (e.g. magnetic and eddy-current separation) unit operations. The recovered materials can be used in metallurgic industry to produce metals with a market value. This option is little effective in increasing the recycling rate of the ELVs (probably in the order of 0.3 – 0.5%). However, by reducing the metals content of ash and slag from ASR treatment, the process could make these waste complying with building materials standards thus increasing the potential for recycling in the construction industry. Since bottom ash and slag from the incineration/gassification of ASR can represent from 4 to 5% of the total weight of ELVs, their recycling is an important step towards the fulfillment of the standards set by the EU Directive mentioned above.

## 6. LANDFILLING

### 6.1 Emission

The ASR, that constitutes about 25% of end of life vehicle, has traditionally been disposed of in landfill (GHK/BIOIS, 2006). Due to its complex matrix and the pollutants content the final deposit requires specific technical and environmental care. The ASR, in fact, may be classified as hazardous waste as could contain dangerous substances in the light fraction and dust (A.A. Zorpas, V.J. Inglezakis, 2012).

One of the parameter to maintain under control is the dissolved organic carbon (DOC) content (Cossu and Lai, 2012). Moreover laboratory studies indicate that insulation foam that may contains fluorocarbons that should be release after disposal in landfill (Scheutz et al., 2010). In different studies also the potential risk linked to the presence of heavy metal (Pb, Zn, Cr, Cd, etc.) was highlight (Kurose et al, 2006; Gonzalez Fernandez et al., 2008); in particular, some metals, such as Pb and Zn, are more presents in the finest fraction while others do not seem linked to a specific grain-size (Gonzalez Fernandez, 2008).

The presence of the both organic and inorganic substances above mentioned may also influence the different recycling processes (Rossetti et al., 2006; Santini 2012).

In order to deal with the leachable fraction of waste that poses a potential threat to the environment the design of landfill should be based on the multibarrier system: decrease in waste quantities, pre-treatment of wastes, final covering and lining systems, control of degradation processes in landfill (Cossu, 1995).

## 6.2 Pre-treatment

The long-term pollution potential of a landfill is determined by the residual concentrations of compounds present in leachate; in order to reduce its environmental impact, a possible pre-treatment option should be represented by the washing of waste before final disposal in landfill. The efficiency of washing pre-treatment was evaluated by using standard batch leaching test by quantifying the improvement of landfill acceptability index calculated as follows:

$$\eta_{A,i} = \frac{e_{r,i} - e_{w,i}}{e_{r,i}} \quad (1)$$

Where:

$e_{r,i}$  = concentration of the  $i$ -substance in the eluate of batch leaching test before washing (BW);  
 $e_{w,i}$  = concentration of the  $i$ -substance in the eluate of batch leaching test after washing (AW).

Batch leaching tests were performed in accordance with standard UNI EN 12547-2, using deionised water as extractant to obtain a liquid to solid ratio of 10 l/kg<sub>TS</sub>. Eluates filtered at 0.45 µm were analysed to determine different parameters: DOC, COD, TKN and metals (As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Se, Zn), chlorides, fluorides, and sulphates. The measured values were compared with limits established by Italian regulations (D.M. 27/09/2010) for waste acceptance in landfill.

The washing pre-treatment was applied at different scale. In the first phase of the experiment the ASR sample was ground to a size < 4 mm by using a laboratory cutting mill (model Retsch SM 2000). The washing test was performed on a laboratory scale by placing 500 g<sub>TS</sub> of sample in an HDPE 5 litre container and adding deionised water to reach different L/S ratios with different treatment times.

The results obtained using a L/S ratio of 5 l/kg<sub>TS</sub> and a duration of 6 h demonstrate a significant reduction of parameters measured in eluate of batch leaching test. In particular DOC concentrations displayed an washing efficiency index ( $\eta_{A,i}$ ) of 53% (concentration decrease in eluate from 102 to 48 mg/l) that was sufficient to enable disposal of waste in a landfill for inert materials (Figure 3). Significant findings were obtained also for metal removal efficiencies; following waste washing, concentrations of copper and zinc in eluates from batch leaching tests were decreased by approximately 90% (Cossu and Lai, 2012).

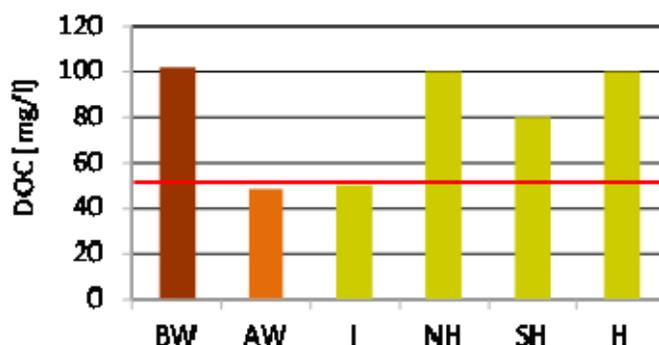


Figure 3. Batch leaching test results for DOC, before (BW) and after washing (AW). compared with the limits for waste acceptance in landfill, set by the Italian regulation (D.M. 27/09/2010), (I: inert; NH: non-hazardous; SH: stable non-reactive hazardous in non-hazardous waste landfills; H: hazardous).

The findings obtained were confirmed by technical scale washing tests ( $L/S=5$ ;  $t=6h$ ), carried out by introducing  $5kg_{TS}$  of material into a mixing device (a commercial mixer for concrete preparation). On average, pre-treatment by washing achieved removal rates of more than 60% for DOC, COD and TKN. Similar removal efficiency (more than 60%) was obtained for metals and anions as attested by EC values (Cossu et al., 2012).

In the last phase of the experiment washing has been applied to ground (size  $< 4$  mm) and not ground waste, in order to verify how the ASR grain size should influence the efficiency of pre-treatment. Preliminary results did not highlight significant differences as the ASR displaying a prevalence of fine fraction (Cossu and Lai, 2012).

Full scale applications need further assessments in order to define the operative costs and the most appropriate operative conditions (washing effluent recirculation and treatment, drying of waste before disposal etc.).

## 8. CONCLUSIONS

The present review paper assessed the ELV and ASR management scenario in Italy. . The available literature was used to fully define the characteristics of ASR, with special emphasis on contaminants. Due to its high heterogeneity and variable composition, ASR is classified as “special waste” or as “hazardous waste” [ ] depending on the presence of hazardous substances such as PCBs, cadmium, and lead. Furthermore, high DOC levels are common in this waste, often preventing its disposal in hazardous waste landfills. Currently, with the exception of a minority fraction that is converted into recyclable material or simply burned, ASR is almost completely disposed in landfills, in Italy as well in most of both EU member and non-member states. This is however in conflict with European Directive 2000/ 53/EC on End-of-Life Vehicles (ELV). It is concluded that, in order to meet the severe targets on reuse and recovery and reuse and recycling ASR must be subjected to some form of recovery, since it constitutes more than 20% of the car mass. The considerable heating value of ASR makes it an interesting feedstock for energy production, which according to the definitions adopted in the EU directives falls in the realm of “recovery”. Car manufacturers recycle many of the materials from wrecked vehicles, but the landfill disposal of the lighter components (Automobile Shredder Residue (ASR), or fluff) is increasingly difficult, and new efforts are necessary to find alternative pathways for fluff disposal. In addition, the ASR waste stream itself is so variable that it cannot be automatically assumed that processes developed for one type of ASR are suitable for other similar waste streams. Despite its appreciable energy content (average Lower Heat Value around 20 MJ/kg), the use of combustion technologies (i.e., co-incineration) to treat ASR may be hampered by the high amounts of combustible material, chlorine, heavy metals, and toxins contained in this waste, as thermal treatment in Waste-to-Energy Plants (WTEP) may result in an increase in acidic gas, heavy metal, and organic pollutant emissions. ASR mixing with other waste streams, such as municipal solid waste, tires or biomass is gradually being applied in commercial processes. Several other solutions have been tested, mainly on a lab or pilot scale, pyrolysis, gasification, injection into a blast furnace and use as a binder in asphalt or as filler in concrete. In the cement industry, generally only low percentages of ASR can be used, unless ASR is previously upgraded and despite remaining problems of increased ash formation, clogging of the fuel injection zone, volatilization of mercury and increased concentrations of hazardous elements in the cement clinker and cement kiln dust. Pyrolysis and gasification are considered emerging technologies although the sole use of ASR is debatable. The environmental impacts of these processes are acceptable, but more supporting data are needed and the advantage over (co-)incineration remains to be proven. However for any process to be viable, its products and services must be able to compete in the marketplace. This means that each

process/plant receiving SR should be characterised by technological robustness and operational flexibility, as well as by the capacity to utilize large quantities of materials. Car manufacturers, shredding companies and SR treatment plants, all operate according to open market conditions and naturally aim to make profit. Should the new technologies enhance the possibility of obtaining energy or materials of significant value, SR suppliers may no longer be willing to pay merely a gate fee, but may request payment for what is now effectively a feedstock material. The car manufacturers could in turn require future ELVs to be processed only at their own facilities, thus bypassing the shredding industry. Under these circumstances no developers of new technologies will be able to obtain adequate financing without providing some indication of a guaranteed and profitable SR feedstock. It is not surprising, therefore, that alternative solutions to landfill have not been diffused on a commercial basis and that it is not clear even to the main stakeholders, which process types they should focus on.

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